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To understand the nature of the superconducting state, we studied the new organic superconductor $(BETS)_2GaCl_4$, that was synthesized by our collaborator L. K. Montgomery. The BETS donor molecule is a variant of the BEDT molecule where four of the inner sulfur atoms are replaced with selenium atoms. The larger selenium atoms produce a larger overlap of the orbitals in the conducting sheets of the crystals and hence produce wider electronic bands.

We used RF penetration depth measurements at 24 MHz initially to map out the H_{c2} diagram of $(BETS)_2GaCl_4$. The measurements were much more successful than we expected yielding many details about the superconducting state and the vortex lattice. As an example, we were able to measure the pinning force potential, often called the Labush parameter, as a function of temperature from 100 mK to 4.5 K. Knowing this parameter enabled us to calculate a critical current at zero temperature of 2×10^4 A/cm^2 for this material.

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Final Report

High and ultra high magnetic field studies of newly synthesized organic superconductors

During the three years of this grant we have increased our knowledge of the nature of the superconducting state in organic superconductors. We have done this by studying the relation of superconductivity and other correlated electron effects to the properties of the carriers in these materials. We have also created a pulsed magnetic field facility capable of creating a 50 tesla (T) field at 0.4 K. This apparatus has been a key component in our studies, and is the largest magnetic field available at an American university. Finally, four papers were published as a result of this work, one is submitted and three are in preparation. In addition, 13 presentations were made at the APS march meetings as a result of this work.

Our goal during this grant was to understand the relation of the crystal structure of novel organic conductors to their electronic properties. The materials we picked were interesting because they are lower dimensional and many have correlated electron ground states such as superconductivity or spin density waves (SDW). Part of our interest was to see if we could understand why some of the materials preferred to be in a SDW state rather than a superconducting state.

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We used RF penetration depth measurements at 24 MHz initially to map out the H_{c2} diagram of $(BETS)_2GaCl_4$. The measurements were much more successful than we expected yielding many details about the superconducting state and the vortex lattice. As an example, we were able to measure the pinning force potential, often called the Labush parameter, as a function of temperature from 100 mK to 4.5 K. Knowing this parameter enabled us to calculate a critical current at zero temperature of 2×10^4 A/cm^2 for this material.

The same measurements can be used to understand the nature of the flux lattice and determine if it has melted or turned to a glassy state. Because the organic systems are clean single crystals they should have small pinning potentials and show flux lattice melting at lower temperatures. From our data we found a flux lattice melting line that is consistent with the two fluid model, but it is not clear if we are really looking at flux lattice melting, or some kind of depinning phenomena. In the future we plan to do very low

field measurements to help discern what we are seeing. This work on the superconducting state in BETS is submitted to Phys. Rev. Lett.

The next two conductors synthesized with the BETS cation by our collaborators were with the $KHg(SCN)_4$ and $NH_4Hg(SCN)_4$ anions. We investigated these two new compounds down to 0.5 K and found no evidence of superconductivity or a SDW state. This is significant because the analogs of these materials made with the ET cation are isostructural and the ET compounds are either superconducting or in a SDW state at low temperatures.

Further investigations showed that the effective masses in the BETS compounds measured by analyzing Shubnikov-de Haas (SdH) oscillations taken in our pulsed fields were significantly lower than in their analog ET compounds. In addition the calculated electron masses in the BETS salts using data from the band structure calculation of Wangbo (NC State), were in good agreement with our measurements.

The difference in the electron effective mass between the BETS and ET compounds suggests that the electron–electron interactions are greater in the ET salts and that the e-e interactions are responsible for the correlated electron states. This in turn suggests that the superconductivity in the ET $NH_4Hg(SCN)_4$ may not be due to only electron phonon interactions as is predicted in conventional superconductors.

A related result can be found in the superconductor (BEDO-TTF)$_2$ReO$_4$ H_2O by subjecting it to isotropic pressure. The cation, BEDO-TTF, in this salt has four of its sulfur atom replaced by oxygen. We measured SdH oscillations in (BEDO-TTF)$_2$ReO$_4$ H_2O up to 27 tesla, at pressures between zero and 14.5 kbar. By taking SdH data at a number of temperatures for each pressure, we were able to determine the effective mass at each pressure. The data shows that the effective mass gradually gets smaller as the pressure increases. Interestingly the Tc of this material goes to zero abruptly at 1.9 kbar. Until band structure calculations are made for different pressures, it is unclear if the decrease in effective mass is due to the change in the band curvature or a change in the e-e interactions.

A second result of this work concerns the two closed Fermi orbits, an electron and a hole pocket one twice the size of the other, that are thought to exist in BEDO. The two frequencies present in the ambient pressure SdH data which correspond to the two pockets are still there and still separated by a factor of two at high pressure (14.5 kbar). It is highly unlikely that both an electron and hole pocket would change size in exactly the same proportion over such immense changes in the pressure. We conclude that only an electron pocket exists at low temperatures (if Hall data is taken into account), and the second frequency is simply a second harmonic of the fundamental which is common to measure in conductors with long scattering times.

This material is now the subject of intense investigation in our lab because we have shown it to have a simple 2-D Fermi surface, and its low Fermi energy allows us to reach the extreme quantum limit in our pulsed

magnetic field. Preliminary results suggest that we may be able to see the quantum Hall effect in this material.

The last subject of discussion is the oldest organic superconductor (TMTSF)ClO_4. In a recent paper Paul Chaikin's group at Princeton has published a complicated phase diagram for this material which includes two order parameters describing spin density waves on different parts of the Fermi surface. The phase diagram is somewhat controversial because it contains a critical point and two lines that have no temperature dependence.

We have discovered a possible new phase line at high fields that may shed light on the validity of this phase diagram. Our data comes from high field transport measurements that peak at 50 T, and we can see evidence of this phase line up to 34 T. Future experiments of the Hall effect will ascertain if the phase line we see fits into a coherent theory for the phase diagram.

Additional experiments were done in pulsed field on a variety of organic conductors with interesting results that are not completely understood yet. The most successful of these was experiments that showed the tunneling between two adjacent Fermi surfaces in an organic superconductor at fields up to 50 T at a temperature of 35 mK. An interesting aspect of this breakdown was that it was modulated by the oscillating density of states on one of the Fermi surfaces due to the SdH oscillations. This work is described on one of the papers below.

Our work over the last three years has used organic conductors to understand the relation between the crystallographic and electron properties of materials. In particular we have learned some of the details that make organic conductors more suitable for superconducting or SDW ground states. To achieve these goals we have built the only pulsed magnetic field laboratory at an American university. This apparatus has become the focal point of our laboratory and will serve our experimental needs for many years to come.

Below is a list of the Papers published as a result of the experiments supported by this grant, and the presentations at professional meetings. Three students will get their Ph. Ds. largely due to this work.

In preparation

S. A. Ivanov, C. H. Mielke, T. J. Coffey, D. A. Howe, C. C. Agosta, L. K. Montgomery and B. Fravel, "Shubnikov-de Haas oscillations in the new organic metals α-$(BETS)_2NH_4Hg(SCN)_4$ and α-$(BETS)_2KHg(SCN)_4$,"

S. A. Ivanov, C. C. Agosta, C. H. Mielke, D. A. Howe, E. B. Yagubskii, N. D. Kushch and S. T. Hannahs, Effective mass and superconductivity in an organic semi-metal under pressure"

D. A. Howe, C. C. Agosta, C. H. Mielke, S. A. Ivanov and T. J. Coffey, "High magnetic field instabilities in a two component spin density wave system."

S. A. Ivanov, C. C. Agosta, D. A. Howe, C. H. Mielke, E. B. Yagubskii, N. D. Kushch, C. Immer and S. T. Hannahs, "Shubnikov-de Haas oscillations in $(BEDO)_2ReO_4H_2O$," Bull. Am. Phys. Soc., **41**, 73, (1996).

D. A. Howe, S. A. Ivanov, C. H. Mielke and C. C. Agosta, "High Magnetic Field Transitions in the Organic Conductor $(TMTSF)_2$ ClO_4," Bull. Am. Phys. Soc., **40**, 266, (1995).

S. A. Ivanov, C. H. Mielke, D. A. Howe, C. C. Agosta, B. K. Ponomarev and B. S. Red'kin, "Magnetic susceptibility of Sm_2 $(MoO_4)_3$ at low temperature," Bull. Am. Phys. Soc., **40**, 779, (1995).

C. H. Mielke, S. A. Ivanov, D. A. Howe, C. C. Agosta and L. K. Montgomery, "Anomalous Susceptibility in the Organic Superconductor λ -$(BETS)_2$ $GaCl_4$," Bull. Am. Phys. Soc., **40**, 804, (1995).

S. A. Ivanov, C. H. Mielke, F. M. Morgan, A. Antia, C. C. Agosta, V. N. Laukhin and E.B. Yagubskii, "New Structure in the Angular and Field Dependencies of the Magnetoresistance of $(BEDT\text{-}TTF)_2TlHg(SCN)_4$," Bull. Am. Phys. Soc., **39**, 879, (1994).

C. H. Mielke, D. A. Howe, S. A. Ivanov, F. M. Morgan, A. Antia, C. C. Agosta and L. K. Montgomery, "Magnetoresistance Measurements of the New Organic Superconductor λ -$(BETS)_2GaCl_4$," Bull. Am. Phys. Soc., **39**, 880, (1994).

D. A. Howe, S. A. Ivanov, C. H. Mielke, F. M. Morgan, A. Antia, C. C. Agosta and L. K. Montgomery, "New Details in the Phase Diagram of $(TMTSF)_2$ ClO_4," Bull. Am. Phys. Soc., **39**, 916, (1994).

C. C. Agosta, J. S. Brooks, P. M. Chaikin and M. Tokumoto, "New High Magnetic Field Phases in Organic Conductors," Bull. Am. Phys. Soc., **38**, 778, (1993).

C. C. Agosta, D. Howe, S. J. Klepper, J. S. Brooks, M. Tokumoto N. Kinoshita and H. Anzai, "Spin Splitting an anomalous High Field Behavior in the Organic Conductor $(BEDT\text{-}TTF)_2KHg(SCN)_4$," Bull. Am. Phys. Soc., **37**, 880, (1992).